

(19)

Eur päisches Patentamt

European Patent Office

Office européen des brevets



(11)

**EP 0 699 521 A1**

(12)

**EUROPEAN PATENT APPLICATION**

published in accordance with Art. 158(3) EPC

(43) Date of publication:

06.03.1996 Bulletin 1996/10

(51) Int. Cl.<sup>6</sup>: **B32B 27/32**, B65D 1/00

(21) Application number: **94907068.4**

(86) International application number: **PCT/JP94/00245**

(22) Date of filing: **17.02.1994**

(87) International publication number:

**WO 95/22456 (24.08.1995 Gazette 1995/36)**

(84) Designated Contracting States:

**AT BE CH DE DK ES FR GB GR IE IT LI LU MC NL PT SE**

(71) Applicant: **OTSUKA PHARMACEUTICAL FACTORY, INC.**

**Naruto-shi Tokushima 772 (JP)**

• **OKAMOTO, Hideshi**

**94-1, Aza-nakazono  
Tokushima 771-11 (JP)**

• **TANIGUCHI, Hirohisa**

**151, Aza-maehama  
Tokushima 772 (JP)**

(72) Inventors:

• **WATANABE, Katsushi**

**68-18, Aza-inamoto  
Tokushima 771-02 (JP)**

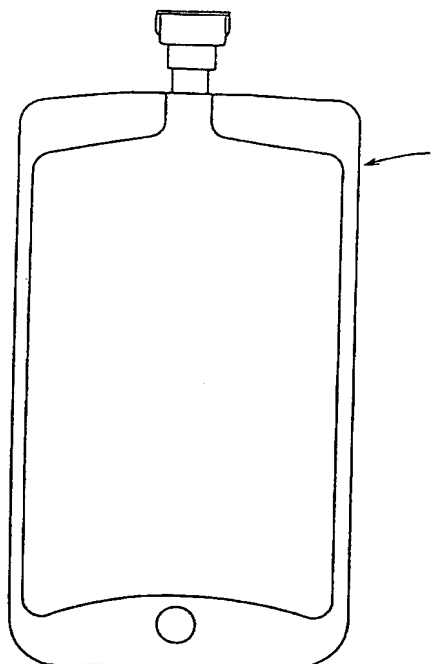
(74) Representative: **Hansen, Bernd, Dr. Dipl.-Chem. et al**

**Hoffmann, Eitle & Partner,  
Patentanwälte,  
Arabellastrasse 4  
D-81925 München (DE)**

**(54) MULTILAYER FILM AND CONTAINER**

(57) A multilayer film according to the present invention is a 5-layer film consisting of ethylene  $\alpha$ -olefin copolymer, and the densities of resin constituting the layers are as follows starting with the outermost: first layer: 0.935-0.950 g/cm<sup>3</sup>, second layer: not more than 0.920 g/cm<sup>3</sup>, third layer: 0.925-0.950 g/cm<sup>3</sup>, fourth layer: not more than 0.920 g/cm<sup>3</sup>, fifth layer: 0.925-0.940 g/cm<sup>3</sup>. A container according to the present invention is molded by using this multilayer film and has high thermal resistance, blocking resistance, tensile strength, flexibility and transparency. Therefore, the multilayer film and container according to the present invention are suitably used for medical purposes.

Fig. 1



**Description**

(Technical Field)

5       The present invention relates to a multilayer film and a container formed by using the same. Specifically, it is used in the medical field, especially as a container for pharmaceutical solution, blood or the like.

(Background Art)

10       As conventional examples of multilayer container for medical use, the following compositions made of polyethylene resin may be listed.

A. Japanese Patent Unexamined Publication No. 62-64363:

15       A three-layer bag composed of linear low density polyethylene, with the density of inner and outer layers being 0.920 g/cm<sup>3</sup> or more and the density of a middle layer being less than 0.920 g/cm<sup>3</sup>.

B. Japanese Patent Unexamined Publication No. 63-248633:

20       A three-layer container composed of linear low density polyethylene, with the density of inner and outer layers being 0.910 to 0.940 g/cm<sup>3</sup> or more, the density of a middle layer being 0.880 to 0.905 g/cm<sup>3</sup>, and a difference in the density between the two being 0.01 g/cm<sup>3</sup> or more.

C. Japanese Patent Unexamined Publication No. 3-277365:

25       A three-layer bag comprising an outer layer of linear low density polyethylene with the density of 0.920 g/cm<sup>3</sup> or more, a middle layer of linear low density polyethylene with the density of 0.915 g/cm<sup>3</sup> or less, and an inner layer of branched low density polyethylene with the density of 0.918 g/cm<sup>3</sup> or more.

30       D. Japanese Patent Unexamined Publication No. 4-266759:

      A bag of three layers or more comprising inner and outer layers composed of a resin in which 5 to 40% of high density polyethylene with the density of 0.945 g/cm<sup>3</sup> or more is mixed with a low density polyethylene having long-chain branches with the density of 0.930 g/cm<sup>3</sup> or less, and a middle layer composed of a resin in which 15% or less of the  
35       same high density polyethylene is mixed with a linear low density polyethylene with the density of 0.920 g/cm<sup>3</sup> or less.

      These compositions, however, have at least one problem out of the followings.

      (1) Since the inner and outer layers are composed of polyethylene resin of low density, heat resistance is insufficient, and seal strength and drop-shock resistance are lowered by high pressure steam sterilization, hot water sterilization,  
40       or other sterilization in high temperature condition.

      (2) Blocking is liable to occur after such sterilization in high temperature condition.

      (3) Wall thickness must be increased in order to enhance the strength of an entire film.

      (4) Since tensile strength is insufficient, the production of bag cannot be speeded up.

      (5) Since the temperature of a heater cannot be increased in heat sealing, it is impossible to seal in a short time.

45       (6) The transparency or flexibility is lowered after sterilization or the like.

      It is hence a primary object of the invention to present a film and a container suited to medical use, eliminating all the above problems.

## (Disclosure of the Invention)

To achieve the above object, a multilayer film of the invention is a five-layer film composed of ethylene- $\alpha$ -olefin copolymer, wherein the density of a resin composing each layer is as follows, in order from an outer layer:

First layer:	0.935 to 0.950 g/cm <sup>3</sup>
Second layer:	0.920 g/cm <sup>3</sup> or less
Third layer:	0.925 to 0.950 g/cm <sup>3</sup>
Fourth layer:	0.920 g/cm <sup>3</sup> or less
Fifth layer:	0.925 to 0.940 g/cm <sup>3</sup>

In the multilayer film of the invention, to the resin of the fifth layer, ethylene- $\alpha$ -olefin copolymer with the density of 0.945 g/cm<sup>3</sup> or more may be added by 10 % by weight or less.

Furthermore, in the multilayer film of the invention, the resin for composing the second layer and fourth layer is preferably:

(1) A mixed resin comprising 40 to 60 % by weight of ethylene- $\alpha$ -olefin copolymer with density of 0.900 to 0.920 g/cm<sup>3</sup>, 40 to 60 % by weight of ethylene- $\alpha$ -olefin elastomer with density of less than 0.900 g/cm<sup>3</sup>, and less than 5 % by weight of high density polyethylene with density of 0.955 g/cm<sup>3</sup> or more,

(2) A mixed resin comprising 20 to 60 % by weight of ethylene- $\alpha$ -olefin copolymer with density of 0.900 to 0.920 g/cm<sup>3</sup>, 40 to 60 % by weight of ethylene- $\alpha$ -olefin elastomer with density of less than 0.900 g/cm<sup>3</sup>, and 30 % by weight or less of ethylene- $\alpha$ -olefin copolymer with density of 0.935 to 0.945 g/cm<sup>3</sup>, or

(3) A mixed resin comprising 20 to 50 % by weight of ethylene- $\alpha$ -olefin copolymer with density of 0.935 to 0.945 g/cm<sup>3</sup>, and 50 to 80 % by weight of ethylene- $\alpha$ -olefin elastomer with density of less than 0.900 g/cm<sup>3</sup>.

As to thickness of each layer, it is preferable that the first layer is 5 to 15%, the second layer is 35 to 45%, the third layer is 3 to 10%, the fourth layer is 35 to 45% and the fifth layer is 5 to 15% of the entire film thickness, respectively.

Every resin composing the film of the invention is an ethylene- $\alpha$ -olefin copolymer, and examples of  $\alpha$ -olefin include propylene, 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene, 1-heptene, 1-octene, 1-nonene, 1-decene, 1-undecene, 1-dodecene, and others with 3 to 12 carbon atoms.

Furthermore, the container for medical use according to the invention is characterized in being formed by using the above multilayer film.

Multilayers film and container according to the invention are improved in heat resistance as compared with conventional products, and are also excellent in blocking resistance, tensile strength, flexibility and transparency.

## (Brief Description of Drawings)

Fig. 1 is a front view of a preferable container according to the invention.

## (Best Mode for Carrying Out the Invention)

Followings are detailed description of resins used in each layer of the film and container of the invention, and a method of manufacturing the same.

## As to First layer:

Since mechanical strength (especially tensile strength) and heat resistance are required, there should be used a resin having the density in a range of 0.935 to 0.950 g/cm<sup>3</sup>, preferably 0.935 to 0.945 g/cm<sup>3</sup>, specifically around 0.940 g/cm<sup>3</sup>. In particular, a resin with the melt flow rate (MFR) of 1.5 to 2.5 g/10 min. (190 °C), and melting point of 120 to 130 °C is more suitable. The thickness should be 5 to 15% of the entire film.

As to Second layer:

In order to make a container have a high flexibility, the ethylene- $\alpha$ -olefin copolymer used in this layer has a density of 0.920 g/cm<sup>3</sup> or less, more preferably 0.890 to 0.905 g/cm<sup>3</sup>. Also, in view of keeping transparency, it is preferable to use a linear copolymer. It is desirable that MFR is 0.5 to 4.0 g/10 min. (190 °C) and melting point is in a range of 105 to 120 °C. For this layer, a resin mixture of two or more types can be used, which preferably is a mixture of 40 to 60 % by weight of resin with density of 0.900 to 0.920 g/cm<sup>3</sup> and 40 to 60 % by weight of ethylene- $\alpha$ -olefin elastomer with density of less than 0.900 g/cm<sup>3</sup>. Furthermore, to this mixed resin, high density polyethylene with density of 0.955 g/cm<sup>3</sup> or more may be added by less than 5 % by weight, or ethylene- $\alpha$ -olefin copolymer with density of 0.935 to 0.945 g/cm<sup>3</sup> may be added by 30 % by weight or less, preferably 20 % by weight or less, so that the heat resistance can be improved without lowering the flexibility. The high density polyethylene may be either  $\alpha$ -olefin copolymer or homopolymer. Other examples of a resin mixture used in this layer include a mixture of 20 to 50 % by weight of ethylene- $\alpha$ -olefin copolymer with density of 0.935 to 0.945 g/cm<sup>3</sup> and a mixture of 50 to 80 % by weight of ethylene- $\alpha$ -olefin elastomer with density of less than 0.900 g/cm<sup>3</sup>. When using a mixed resin, the density of the entire resin needs to be set within the above specified range. The thickness of the layer should be 35 to 45% of the entire film.

As to Third layer:

To keep the strength (tenacity) of the entire film, a resin of which density is in a range of 0.925 to 0.950 g/cm<sup>3</sup>, preferably 0.925 to 0.940 g/cm<sup>3</sup> is used in the middle layer. The resin may be in either linear or branched, but a linear polymer is preferable in order to improve an affinity for other layers and to keep transparency. It is preferable that MFR is in a range of 1.5 to 2.5 g/min. (190 °C) and melting point is in a range of 120 to 130 °C.

As a resin used in this layer, a mixed resin comprising two types of ethylene- $\alpha$ -olefin copolymer may be used, but in such a case, a resin having a density of 0.935 to 0.945 g/cm<sup>3</sup> should be contained at least by 40 to 60 % by weight, and then, the other proper ethylene- $\alpha$ -olefin copolymer will be selected so that the density after blending may fall within the above specified range.

The thickness is approximately in a range of 3 to 10% of the entire film.

As to Fourth layer:

The same resin as used in the second layer is disposed, thus maintaining flexibility of a container. The layer thickness can be selected out of the range defined in the second layer.

As to Fifth layer:

A resin having blocking resistance and heat resistance is disposed, thus making it possible to withstand sterilization of high temperature, to prevent blocking and to carry out heat seal easily. Specifically, a resin having a density of 0.925 to 0.940 g/cm<sup>3</sup> should be used. The same resin as used in the third layer can be used, but in order to increase heat resistance, a resin with density of 0.945 g/cm<sup>3</sup> or more can be added up to about 10 % by weight. In such a case, density of the entire resin have to be set in the above specified range. The thickness should be 5 to 15% of the entire film.

As a method of manufacturing a film of the invention, there are water-cooled or air-cooled extrusion inflation method, co-extrusion T-die method, dry lamination method, extrusion lamination method, and others. In particular, the water-cooled co-extrusion inflation method and co-extrusion T-die method are preferable from the viewpoint of performance, in particular, transparency, economy and sanitation. In either method, it is necessary to perform at a temperature for melting the resin of each layer, but if the temperature is raised too high, part of the resin is thermally decomposed, and the performance may be lowered due to decomposition products. Therefore, the temperature condition in manufacturing the film of the invention is normally 150 to 250 °C, preferably 170 to 200 °C. In order to maintain transparency, the resin comprising each layer may have a minimum difference in MFR.

The thickness of the film of the invention manufactured in this method is generally 100 to 300 $\mu$ , which may be increased or decreased properly depending on the purpose of use, and a sufficient strength can be kept even at a thickness of about 200 $\mu$ .

According to the method of manufacturing a container in the invention, a film in the form of tube or sheet obtained in the above described manner, is cut and heat-sealed by the conventional method, and a mouth part or the like is attached by heat seal or other means, thus manufacturing a container having a specified shape and dimensions. As the heat seal condition for the film, the temperature may be raised to about 120 to 160 °C, and for a film with thickness of, for example, about 200 $\mu$ , it is possible to seal in a short time of 0.5 to 3 seconds in the above temperature range. Linear low density polyethylene is suitable for the mouth part because it fuses well with the inner layer of the film of the invention.

(Example)

An evaluation experiment of a container formed by using the multilayer film of the invention will be described below.

First, laminate sheets of synthetic resin and layer constitution shown in Table 1, were formed by water-cooled co-extrusion inflation method. Using these products, a container 1 for medical use having a capacity of 500 ml as shown in Fig. 1 was formed. Heat sealing for the film when forming the container was done for 2 seconds at 140 °C.

Then, in the obtained container 1 for medical use, the characteristics were evaluated according to the following method. That is, heat resistance was measured by visually observing the state of the container in view of deformation, breakage and seal leak after filling with internal solution and sterilizing in high pressure steam for 40 minutes at 110 °C.

Drop-shock test was conducted by cooling at about 4 °C, falling the container five times each from three directions from a height of 1.2 m, thus visually observing breakage and seal leak.

Flexibility was evaluated by visually observing a natural discharge property of the internal solution.

Transparency was evaluated by visually observing the container after filling with distilled water and sterilizing in high pressure steam, and further by measuring transmission rate of 450 nm light.

In addition, by visually observing the entire appearance of the container, crease, blocking, deformation, and breakage were investigated.

On the other hand, as comparative examples, laminate sheets of synthetic resin and layer constitution shown in Table 2 were formed in the same manner as described above, and using these products, containers for medical use were formed in the same procedure. Then, each of the characteristics was evaluated.

In Tables 1 and 2, the following abbreviations are used.

PE-1 = linear medium density polyethylene (ethylene-1-butene copolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.940 g/cm<sup>3</sup>, MFR = 2.1 g/10 min (190 °C)]

PE-2 = linear low density polyethylene (ethylene-1-butene copolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.900 g/cm<sup>3</sup>, MFR = 0.9 g/10 min (190 °C)]

PE-3 = linear medium density polyethylene (ethylene-1-butene copolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.930 g/cm<sup>3</sup>, MFR = 2.1 g/10 min.(190 °C)]

PE-4 = PE-3 blended with 5 % by weight of high density polyethylene (ethylene-1-butene copolymer) [made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.956 g/cm<sup>3</sup>, MFR = 0.9 g/10 min.(190 °C)] <density of the mixed resin = 0.931 g/cm<sup>3</sup>>

PE-5 = a mixed resin <density of the mixed resin = 0.905 g/cm<sup>3</sup>> comprising:

45.5 % by weight of linear low density polyethylene (ethylene-1-butene copolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.920 g/cm<sup>3</sup>, MFR = 2.1 g/10 min.(190 °C)],

50 % by weight of ethylene-1-butene elastomer

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.885 g/cm<sup>3</sup>, MFR = 0.5 g/10 min.(190 °C)],

and 4.5 % by weight of high density polyethylene (homopolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.965 g/cm<sup>3</sup>, MFR = 15 g/10 min.(190 °C)]

PE-6 = a mixed resin <density of the mixed resin = 0.904 g/cm<sup>3</sup>> comprising:

47.5 % by weight of linear low density polyethylene (ethylene-1-butene copolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.920 g/cm<sup>3</sup>, MFR = 2.1 g/10 min.(190 °C)],

50 % by weight of ethylene-1-butene elastomer

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.885 g/cm<sup>3</sup>, MFR = 0.5 g/10 min.(190 °C)],

and 2.5 % by weight of high density polyethylene (homopolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.965 g/cm<sup>3</sup>, MFR = 15 g/10 min.(190 °C)]

PE-7 = a mixed resin <density of the mixed resin = 0.898 g/cm<sup>3</sup>> comprising:

45.5 % by weight of linear low density polyethylene (ethylene-1-butene copolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.905 g/cm<sup>3</sup>, MFR = 2.9 g/10 min.(190 °C)],

50 % by weight of ethylene-1-butene elastomer

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.885 g/cm<sup>3</sup>, MFR = 0.5 g/10 min.(190 °C)],

and 4.5 % by weight of high density polyethylene (homopolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.965 g/cm<sup>3</sup>, MFR = 15 g/10 min.(190 °C)]

PE-8 = a mixed resin <density of the mixed resin = 0.905 g/cm<sup>3</sup>> comprising:

40 % by weight of linear low density polyethylene (ethylene-1-butene copolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.920 g/cm<sup>3</sup>, MFR = 2.9 g/10 min.(190 °C)],

50 % by weight of ethylene-1-butene elastomer

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.885 g/cm<sup>3</sup>, MFR = 0.5 g/10 min.(190 °C)],

and 10 % by weight of PE-1

PE-9 = a mixed resin <density of the mixed resin = 0.904 g/cm<sup>3</sup>> comprising:

45 % by weight of linear low density polyethylene (ethylene-1-butene copolymer)

## EP 0 699 521 A1

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.920 g/cm<sup>3</sup>, MFR = 2.9 g/10 min.(190 °C)],  
50 % by weight of ethylene-1-butene elastomer

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.885 g/cm<sup>3</sup>, MFR = 0.5 g/10 min.(190 °C)],  
and 5 % by weight of PE-1

PE-10 = 1:1 mixed resin <density of the mixed resin = 0.930 g/cm<sup>3</sup>> comprising:

linear low density polyethylene (ethylene-1-butene copolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.920 g/cm<sup>3</sup>, MFR = 2.9 g/10 min.(190 °C)],  
and PE-1

PE-11 = a linear low density polyethylene (ethylene-1-butene copolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.925 g/cm<sup>3</sup>, MFR = 2.3 g/10 min.(190 °C)]

PE-12 = a mixed resin <density of the mixed resin = 0.907 g/cm<sup>3</sup>> comprising:

30 % by weight of linear low density polyethylene (ethylene-1-butene copolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.920 g/cm<sup>3</sup>, MFR = 2.9 g/10 min.(190 °C)],  
50 % by weight of ethylene-1-butene elastomer

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.885 g/cm<sup>3</sup>, MFR = 0.5 g/10 min.(190 °C)],  
and 20 % by weight of PE-1

PE-13 = a mixed resin <density of the mixed resin = 0.909 g/cm<sup>3</sup>> comprising:

20 % by weight of linear low density polyethylene (ethylene-1-butene copolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.920 g/cm<sup>3</sup>, MFR = 2.9 g/10 min.(190 °C)],  
50 % by weight of ethylene-1-butene elastomer

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.885 g/cm<sup>3</sup>, MFR = 0.5 g/10 min.(190 °C)],  
and 30 % by weight of PE-1

PE-14 = a mixed resin <density of the mixed resin = 0.902 g/cm<sup>3</sup>> comprising:

70 % by weight of ethylene-1-butene elastomer

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.885 g/cm<sup>3</sup>, MFR = 0.5 g/10 min.(190 °C)],  
and 30 % by weight of PE-1

PE-15 = a mixed resin <density of the mixed resin = 0.907 g/cm<sup>3</sup>> comprising: 60 % by weight of ethylene-1-butene  
elastomer

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.885 g/cm<sup>3</sup>, MFR = 0.5 g/10 min.(190 °C)],  
and 40 % by weight of PE-1

PE-16 = a linear low density polyethylene (ethylene-1-butene copolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.920 g/cm<sup>3</sup>, MFR = 2.9 g/10 min.(190 °C)]

PE-17 = a high density polyethylene (homopolymer)

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.965 g/cm<sup>3</sup>, MFR = 15 g/10 min.(190 °C)]

PE-18 = an ethylene-1-butene elastomer

[made by MITSUI PETROCHEMICAL INDUSTRIES, LTD.; density = 0.885 g/cm<sup>3</sup>, MFR = 0.5 g/10 min.(190 °C)]

EP 0 699 521 A1

Symbols in the tables denotes that ⊙ is excellent, ○ is good, △ is slightly poor, and x is poor.

Table 1

No.	Outer Layer      ←      Inner Layer					Total thickness (μ)	Heat resistance	Drop-shock test	Flexibility	Transparency		Appearance	Overall evaluation
	Kind of resin [thickness (μ)]									Visual	Transmission (%)		
1	PE-1 (15)	PE-2 (80)	PE-3 (10)	PE-2 (80)	PE-3 (15)	200	⊙	⊙	⊙	⊙	90.6	⊙	⊙
2	PE-1 (15)	PE-2 (95)	PE-3 (10)	PE-2 (95)	PE-3 (15)	230	⊙	⊙	⊙	⊙	89.3	⊙	⊙
3	PE-1 (15)	PE-2 (80)	PE-3 (10)	PE-2 (80)	PE-4 (15)	200	⊙	⊙	⊙	⊙	90.3	⊙	⊙
4	PE-1 (15)	PE-5 (80)	PE-3 (10)	PE-5 (80)	PE-3 (15)	200	⊙	⊙	⊙	⊙	85.8	⊙	⊙
5	PE-1 (15)	PE-6 (80)	PE-3 (10)	PE-6 (80)	PE-3 (15)	200	⊙	⊙	⊙	⊙	86.5	⊙	⊙
6	PE-1 (15)	PE-7 (80)	PE-3 (10)	PE-7 (80)	PE-3 (15)	200	⊙	⊙	⊙	⊙	87.0	⊙	⊙
7	PE-1 (15)	PE-8 (80)	PE-3 (10)	PE-8 (80)	PE-3 (15)	200	⊙	⊙	⊙	⊙	85.1	⊙	⊙
8	PE-1 (15)	PE-5 (80)	PE-3 (10)	PE-5 (80)	PE-3 (15)	200	⊙	⊙	⊙	⊙	85.1	⊙	⊙
9	PE-1 (15)	PE-9 (80)	PE-3 (10)	PE-9 (80)	PE-3 (15)	200	⊙	⊙	⊙	⊙	85.7	⊙	⊙
10	PE-1 (15)	PE-8 (80)	PE-10 (10)	PE-8 (80)	PE-3 (15)	200	⊙	⊙	⊙	⊙	85.1	⊙	⊙

Table 1(continued)

No.	Outer Layer      ←      Inner Layer					Total thickness ( $\mu$ )	Heat resistance	Drop-shock test	Flexibility	Transparency		Appearance	Overall evaluation
	Kind of resin [thickness ( $\mu$ ) ]									Vis-ual	Trans-mission (%)		
11	PE-1 (15)	PE-8 (80)	PE-3 (10)	PE-8 (80)	PE-11 (15)	200	⊙	⊙	⊙	⊙	87.5	⊙	⊙
12	PE-1 (15)	PE-12 (80)	PE-3 (10)	PE-12 (80)	PE-3 (15)	200	⊙	⊙	⊙	⊙	84.6	⊙	⊙
13	PE-1 (15)	PE-13 (80)	PE-3 (10)	PE-13 (80)	PE-3 (15)	200	⊙	⊙	⊙	⊙	83.9	⊙	⊙
14	PE-1 (15)	PE-14 (80)	PE-3 (10)	PE-14 (80)	PE-3 (15)	200	⊙	⊙	⊙	⊙	83.2	⊙	⊙
15	PE-1 (15)	PE-15 (80)	PE-3 (10)	PE-15 (80)	PE-3 (15)	200	⊙	⊙	⊙	○	81.3	⊙	⊙



Table 2

No.	Outer Layer ← Inner Layer					Total thickness (μ)	Heat resistance	Drop-shock test	Flexibility	Transparency		Appearance	Overall evaluation
	Kind of resin [thickness (μ)]									Vis-ual	Trans-mission (%)		
I	PE-16 (15)	PE-2 (80)	PE-3 (10)	PE-2 (80)	PE-3 (15)	200	△	⊙	⊙	△	86.5	△	△
II	PE-1 (15)	PE-2 (80)	PE-3 (10)	PE-2 (80)	PE-17 (15)	200	⊙	×	○	○	82.9	○	×
III	PE-1 (15)	PE-3 (80)	PE-1 (10)	PE-3 (80)	PE-1 (15)	200	⊙	△	○	○	81.5	○	△
IV	PE-1 (15)	PE-2 (80)	PE-3 (10)	PE-2 (80)	PE-18 (15)	200	×	△	⊙	○	81.0	○	×

It is clear from Table 1 and Table 2 that the films and containers of the invention are excellent in flexibility, heat resistance, strength, and transparency.

#### (Industrial Applicability)

The multilayer film and container according to the invention possess many advantages, for example:

- \* Capable of withstanding high pressure steam sterilization, hot water sterilization, or other high temperature sterilization.
- \* Capable of reducing the overall thickness of a film.
- \* Excellent in tensile strength and capable of producing a bag at high speed.
- \* Hardly inducing blocking.
- \* Capable of conducting heat sealing at high temperature and in short time, and excellent in sealing performance.
- \* Capable of maintaining excellent flexibility and transparency even after sterilization.

Hence it can be preferably used as container for medical use such as transfusion bag, blood bag or the like.

## Claims

1. A multilayer film, being a five-layer film composed of ethylene- $\alpha$ -olefin copolymer, wherein the density of a resin composing each layer is as follows, in order from the outer layer:

First layer:	0.935 to 0.950 g/cm <sup>3</sup>
Second layer:	0.920 g/cm <sup>3</sup> or less
Third layer:	0.925 to 0.950 g/cm <sup>3</sup>
Fourth layer:	0.920 g/cm <sup>3</sup> or less
Fifth layer:	0.925 to 0.940 g/cm <sup>3</sup>

2. A multilayer film according to claim 1, wherein in the resin of the fifth layer, an ethylene- $\alpha$ -olefin copolymer with the density of 0.945 g/cm<sup>3</sup> or more is blended by 10 % by weight or less.

3. A multilayer film according to claim 1, wherein a resin used in the second and fourth layers is a mixed resin comprising 40 to 60 % by weight of ethylene- $\alpha$ -olefin copolymer with density of 0.900 to 0.920 g/cm<sup>3</sup>, 40 to 60 % by weight of ethylene- $\alpha$ -olefin elastomer with density of less than 0.900 g/cm<sup>3</sup>, and less than 5 % by weight of high density polyethylene with density of 0.955 g/cm<sup>3</sup> or more.

4. A multilayer film according to claim 1, wherein a resin used in the second and fourth layers is a mixed resin comprising 20 to 60 % by weight of ethylene- $\alpha$ -olefin copolymer with density of 0.900 to 0.920 g/cm<sup>3</sup>, 40 to 60 % by weight of ethylene- $\alpha$ -olefin elastomer with density of less than 0.900 g/cm<sup>3</sup>, and 30 % by weight or less of ethylene- $\alpha$ -olefin copolymer with density of 0.935 to 0.945 g/cm<sup>3</sup>.

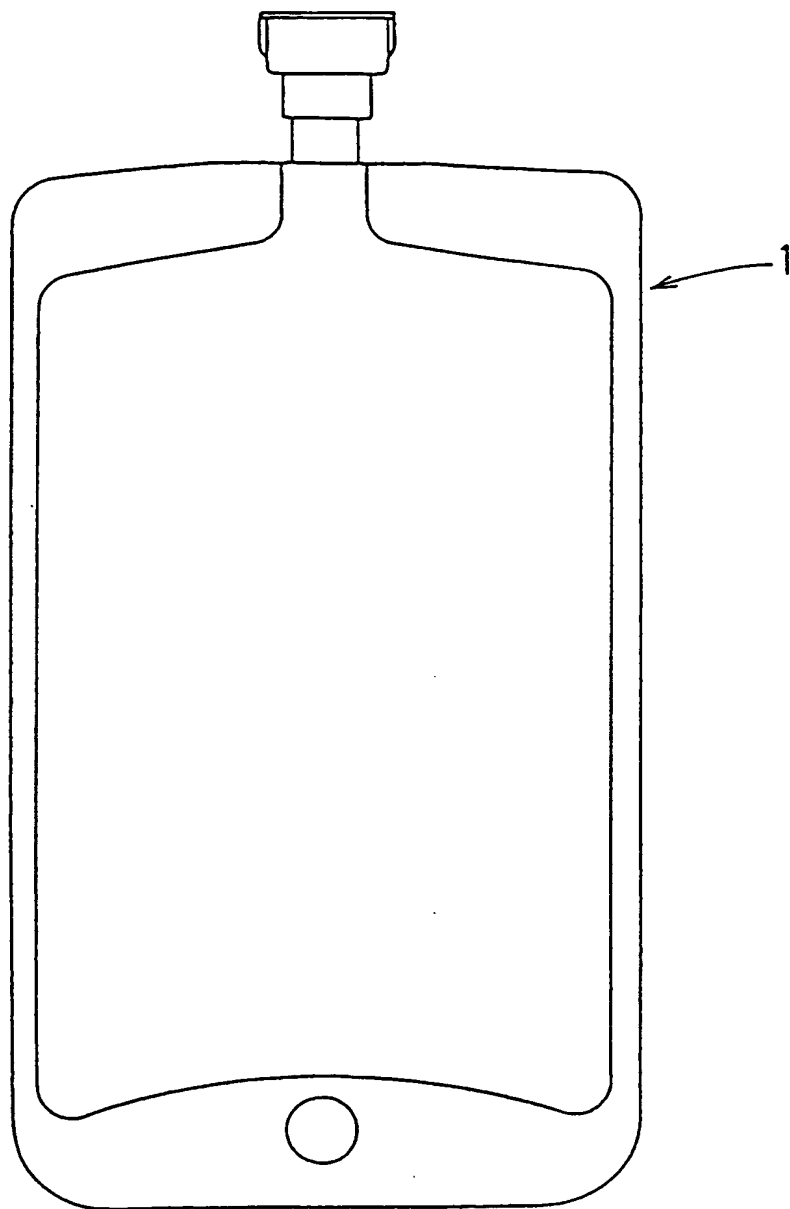
5. A multilayer film according to claim 1, wherein a resin used in the second and fourth layers is a mixed resin comprising 20 to 50 % by weight of ethylene- $\alpha$ -olefin copolymer with density of 0.935 to 0.945 g/cm<sup>3</sup>, and 50 to 80 % by weight of ethylene- $\alpha$ -olefin elastomer with density of less than 0.900 g/cm<sup>3</sup>.

6. A multilayer film according to any one of claims 1 to 5, wherein thickness of each layer for the entire thickness of the film lies in the following range.

First layer	5 to 15%
Second layer	35 to 45%
Third layer	3 to 10%
Fourth layer	35 to 45%
Fifth layer	5 to 15%

7. A container formed by using a multilayer film according to any one of claims 1 to 6.

Fig. 1



## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP94/00245

## A. CLASSIFICATION OF SUBJECT MATTER

Int. Cl<sup>5</sup> B32B27/32, B65D1/00

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Int. Cl<sup>5</sup> B32B27/00-27/32, B65D1/00, 65/40, A61J1/00

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Jitsuyo Shinan Koho

1926 - 1994

Kokai Jitsuyo Shinan Koho

1971 - 1994

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP, A, 62-64363 (Showa Denko K.K.), March 23, 1987 (23. 03. 87), Claim and example 3 & EP, A, 216509 & US, A, 4775562	1-7
A	JP, A, 63-248633 (Kyoraku K.K.), October 14, 1988 (14. 10. 88), Claim, (Family: none)	1-7
A	JP, A, 4-40956 (Showa Denko K.K.), February 12, 1992 (12. 02. 92), Claim, (Family: none)	1-7
A	JP, A, 4-266759 (Showa Denko K.K.), September 22, 1992 (22. 09. 92), Claim, (Family: none)	2
A	JP, A, 58-216061 (Terumo Corp.), December 15, 1983 (15. 12. 83), Claim, (Family: none)	3-5

☐ Further documents are listed in the continuation of Box C.☐ See patent family annex.

\* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"Z" document member of the same patent family

Date of the actual completion of the international search

April 26, 1994 (26. 04. 94)

Date of mailing of the international search report

May 17, 1994 (17. 05. 94)

Name and mailing address of the ISA/

Japan se Patent Office

Authorized officer

Facsimile No.

Telephone No.